On-Site Inspection RadioIsotopic Spectroscopy (OSIRIS) System-Integration and Field-Testing Report

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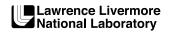
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Abstract

e have designed a system for the acquisition and analysis of high-resolution gamma-ray spectra during on-site inspections under the Comprehensive Nuclear-Test-Ban Treaty (CTBT).

The On-Site Inspection RadioIsotopic Spectroscopy—OSIRIS —gamma-ray spectrometer is a mechanically-cooled, battery-powered ORTEC IDM-200V, chosen to avoid the need for liquid nitrogen during on-site inspections. The spectrometer is controlled and the spectral data analyzed by a ruggedized notebook computer. Spectral data analysis is similar to that performed by traditional gamma-ray spectrometers, except the OSIRIS gamma-ray spectral data are filtered to display only radioisotopic information relevant to CTBT on-site inspections, for example, the fission-product isotope zirconium-96 (Zr-95) and its daughter isotope Nb-95.

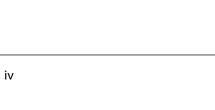
The OSIRIS software assists the user during operation with software checklists for startup and shutdown, and menu-selected display windows during energy calibration and data acquisition. OSIRIS can be operated in either sample-counting mode or the *in-situ* mode.

In this report, the extensive system-integration tests of the Osiris system carried out in 2015-2017 are described, including tests at Idaho National Laboratory, at the Nevada National Security Site (formerly, the Nevada Test Site), and at Pacific Northwest National Laboratory in Richland, Washington. We also note recent software enhancements and the steps to our successful commercialization of Osiris.



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1. Introduction

The On-Site Inspection RadioIsotopic Spectroscopy (OSIRIS) system is a special-purpose spectrometer for on-site inspections under the Comprehensive Nuclear-Test-Ban Treaty (CTBT). [1] The OSIRIS high-resolution spectrometer is controlled by a ruggedized notebook computer running custom data acquisition and data analysis software. The OSIRIS spectrometer and computer are shown in Figure 1.

The spectrometer can be used to identify radioisotopes in samples collected from the soil or, in the "in-situ" counting geometry, it can directly measure radioisotopes deposited on the ground. In either geometry, the spectrometer measures the energy-intensity pattern or spectrum of gamma rays striking the spectrometer germanium crystal. From the energies of the gamma-ray peaks in the

spectrum, the emitting radioisotopes are determined, and from the intensities of the peaks, the quantities of each radioisotope can be learned. A fission-product gamma-ray energy spectrum is shown in Figure 2.

In this report, we review the final Osiris hardware and software designs and the extensive Osiris systems tests conducted from 2015 through 2017.

1.1 CTBT On-Site Inspection Requirements

Under the CTBT, each State Party has the right to request an on-site inspection. The treaty and its Protocol allow the use of gamma-ray spectroscopy during on-site inspection, and they also allow for measurement restrictions as a managed access tool by the Inspected State Party. [2]

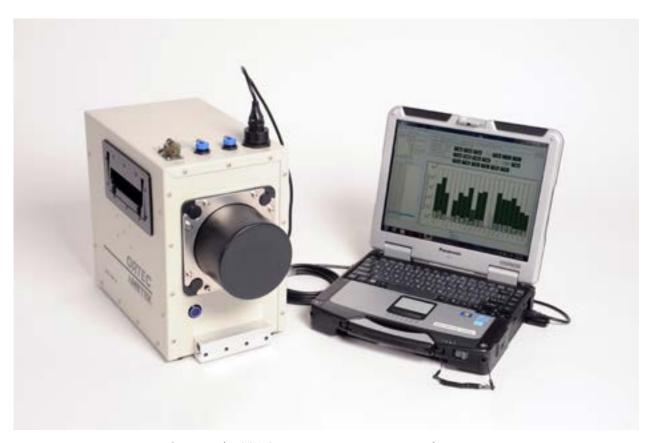


Figure 1. The OSIRIS gamma-ray spectrometer and computer.

An underground nuclear explosion releases noble gases like the radioxenon isotopes by diffusion through cracks in the rocks, and specialized instruments for on-site noble gas collection and analysis have been developed. [3] If an underground test vents, other radioisotopes may be deposited on the ground as particulate matter, like cesium-137 (Cs-137) and these isotopes are best identified by a gamma-ray spectroscopy system like OSIRIS.

Paragraph 89(b) of the CTBT Protocol limits the radiation measurements to those isotopes and energies relevant to the determination that a nuclear explosion has occurred so as not to disclose irrelevant and potentially proliferation-sensitive information. [4] To minimize the intrusiveness of gamma-ray spectral measurements during on-site inspections, Working Group B of the CTBT Organization Preparatory Commission has agreed to restrict gamma-ray spectral measurements during a CTBT on-site inspection to the seventeen activation and fission products listed in Table 1. [5]

1.2 Osiris Design Choices

An "off-the-shelf" commercial software package would be sufficient for CTBT on-site gamma-ray spectroscopy, except it may reveal the gamma-ray peaks of other radioisotopes beyond the seventeen listed in Table 1. In addition, all of the commercial spectroscopy packages we are aware of display actual spectra to users during data acquisition and analysis. An experienced spectroscopist can identify the gamma-ray peaks of other radioisotopes visually, even when not flagged by the software.

Instead of commercial software, we adopted and modified the data acquisition and analysis software of the Portable Isotopic Neutron Spectroscopy (PINS) prompt gamma-ray neutron activation analysis system [6] for OSIRIS. PINS was developed at Idaho National Laboratory for on-site inspections under the Chemical Weapons

Table 1. The CTBT-relevant fission-product isotopes and their half-lives*

Isotope	Half-life	Parent
Ba-140	12.79 days	
Ce-141	32.52 days	
Ce-144	284.89 days	
Cs-134	2.062 years	
Cs-137	30.07 years	
I-131	8.02 days	
I-132	2.295 hours	3.204-day Te-132
La-140	1.678 days	12.79-day Ba-140
Mo-99	2.748 days	
Nb-95	34.97 days	64.0-day Zr-95
Nd-147	10.98 days	
Pr-144	17.28 minutes	284.89-day Ce-144
Rh-106	29.8 sec	373.6-day Ru-106
Ru-103	39.26 days	
Tc-99m	6.01 hours	2.748-day Mo-99
Te-132	3.204 days	
Zr-95	64.02 days	

^{*}R.B. Firestone, Editor, *Table of Isotopes, Eighth Edition* (New York: Wiley, 1996)

Convention. The PINS software optionally can hide gamma-ray spectra from users, presenting only summary information. For OSIRIS, hiding gamma-ray spectra is not optional—it is mandatory, and instead of displaying spectra like Figure 2, the display presents only summary information on CTBT-relevant radioisotopes, as shown in Figure 3.

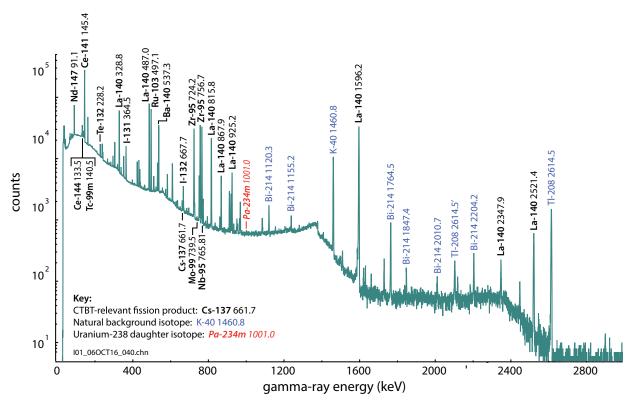


Figure 2. Fission-product gamma-ray energy spectrum, measured from a uranium sample irradiated in the INL Advanced Test Reactor.

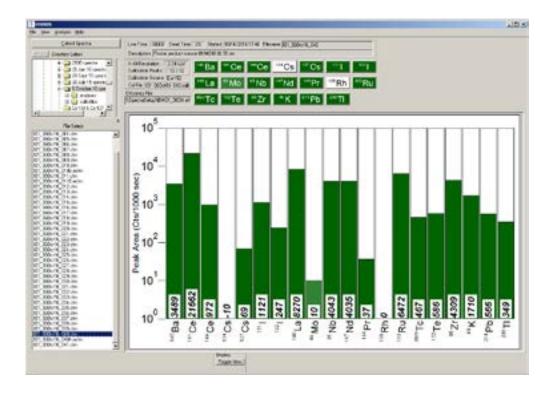


Figure 3. Osiris main radioisotope display

The complexity of fission-product gamma-ray spectra requires the use of a high-purity germanium (HPGe) detector to resolve closely-spaced gamma-ray peaks. It is likely that on-site inspections under the CTBT will take place in remote locations, where access to liquid nitrogen and even electricity may be limited. Accordingly, we have chosen a battery-powered, mechanically-cooled HPGe gamma-ray spectrometer also used in some versions of the PINS system.

1.3 Technology Readiness Level Goal

The Osiris project goal is the development of a commercially-available gamma-ray spectrometer, at Technology Readiness Level (TRL) 8. [7] At the beginning of Osiris development, the hardware and software components adapted from PINS were at TRL- 9, corresponding to systems that have been used successfully in military field applications.

1.4 Performance Metrics

In addition to restricting spectral information to the seventeen CTBT-relevant isotopes, the following metrics are used to evaluate OSIRIS system performance:

- Detection of the CTBT-relevant fission-product isotopes actually present in a spectrum (high true-positive fraction).
- Non-detection of the CTBT-relevant fissionproduct isotopes not present in a spectrum (low false-positive fraction).
- Correct areas for the CTBT-relevant peaks truly present in the spectrum. The radioactivity, in becquerels or microcuries, of an isotope is directly proportional to its gamma-ray peak area.
- Accurate spectrometer energy calibrations.
- Stable spectrometer electronic gain between energy calibrations.

2. Osiris Hardware

The principal Osiris hardware components are an HPGe gamma-ray spectrometer and a notebook computer for control, data acquisition, and data analysis.

2.1 Prototype Gamma-Ray Spectrometer

The ORTEC Transpec-100 gamma-ray spectrometer was selected as the Osiris prototype because of its proven reliability, freedom from liquid nitrogen supplies, battery operation, and 40% relative-efficiency Ge detector. The Transpec-100 and the nearly identical ORTEC Detective-100 [8] gamma-ray spectrometers have been used in U.S. military

applications since 2005. The Transpec-100 Ge crystal is cooled to about 100 Kelvin (–173 °C, –279 °F.) by an internal electrically-powered refrigerator.

The Transpec-100 lithium-ion battery can power the instrument for 12 hours. The Transpec-100 also includes a built-in digital-signal-processing multichannel analyzer.

The Transpec-100 was used at the Comprehensive Nuclear-Test-Ban Treaty Organization Integrated Field Exercise (IFE14) conducted in Jordan during November-December 2014. [9] The spectrometer was used in the *in-situ* tripod-mounted counting geometry during IFE14, as shown in Figure 4.



Figure 4. The Transpec-100 gamma-ray spectrometer is set up for in-situ geometry counting in Jordan during the IFE14 CTBTO exercise.

2.2 Gamma-Ray Spectrometer Upgrade

A modified ORTEC IDM-200-V HPGe spectrometer, displayed in Figure 5, replaced the Transpec-100 in 2016 to upgrade spectral data security. Like the Transpec-100, its energy resolution at 1332 keV is 2.3 keV FWHM or less, and its relative efficiency is higher, 50%. Both the Transpec-100 and the IDM employ the same digital-signal-processing electronics.

However, a standard IDM retains the most recent gamma-ray spectrum in internal memory, even when powered off, and available for download on another computer. To preclude this mode of data loss, Osiris IDM spectrometers have been factory modified to erase the gamma-ray spectrum in internal memory whenever the USB cable is disconnected from the control computer.

The IDM spectrometer features are summarized in Table 2.

2.3 Control Computer

The OSIRIS control computer is a ruggedized notebook computer, running the Microsoft Windows operating system. It communicates with the spectrometer via USB cable. For data security reasons, this cable must not be disconnected while the spectrometer is counting gamma rays.

The computer controls the spectrometer. It also analyzes, displays, and stores gamma-ray spectral data, as explained below.

2.4 Auxiliary Equipment

The OSIRIS systems also include a tripod for measurements using the *in-situ* geometry, shown above in Figure 4, a source holder for sample measurements, shown in Figure 6, military-grade external lithium batteries and charger for field operation away from AC line power, and three custom foam-lined shipping containers.

Table 2. OSIRIS gamma-ray spectrometer features [10]

Table 21 Oshiis ganiina ray speed	official reactives [10]
Model:	ORTEC Interchangeable Detector Module IDM-200-V
HPGe crystal material:	p-type
Relative efficiency:	50%
Energy resolution:	< 2.3 keV @ 1332 keV FWHM
Multichannel analyzer (MCA):	built-in digital-signal-processing MCA, 16K channels
Dimensions:	16.3 x 13.4 x 8.4 inches (41.3 x 34.0 x 21.2 cm)
Weight:	39 pounds (18 kilograms)
Input power:	30 watts when cold
Internal battery life:	2.5 hours
External battery:	extends operating life to 8-10 hours; hot-swappable
Cool-down time:	15 hours from room temperature



Figure 5. ORTEC IDM-200-V gamma-ray spectrometer and external battery.



Figure 6. IDM spectrometer and source holder

3. Osiris Software

3.1 Data Acquisition Software

The spectrometer is controlled from the computer using ORTEC CONNECTIONS low-level commands. However, users never see that complexity, and the graphical-user interface guides the operator through startup, data collection, and shutdown.

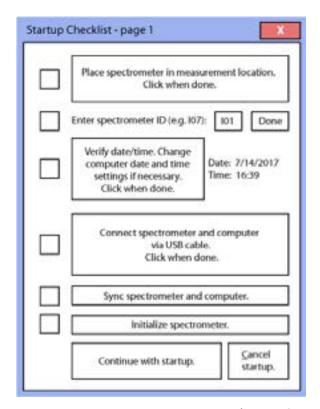
At startup, the user-friendly checklists guide the operator through the initialization steps, in proper order, as shown in Figure 7. Initialization includes setting multichannel analyzer parameters, enabling detector high voltage, and performing a spectrometer pole-zero cancellation. Next the operator is instructed to calibrate the spectrometer energy and peakwidth scales, using a gamma-ray check source, typically Eu-152.

Prior to spectral data collection, the operator presets the measurement live time. Controls are provided to start and stop data collection, and to extend live time, if desired. During data acquisition, the spectrum is analyzed and the information display is updated every 10 seconds. At the end of data acquisition, the raw spectrum is encrypted, as explained in section 3.6, and saved along wih an open (non-encrypted) summary data analysis file.

The spectrometer is shutdown via another checklist.

3.2 Spectral Analysis

Spectroscopy is the art of measuring and interpreting wavelength-intensity or energy-intensity patterns, or "spectra."



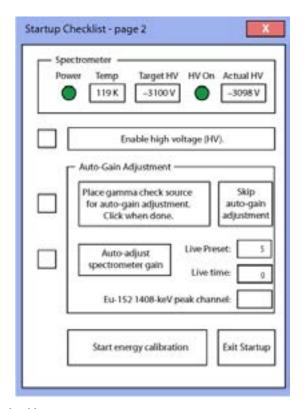


Figure 7. Startup checklist pages.

Each Osiris spectrum, like that displayed in Figure 2, may contain over 100 vertical spikes or peaks, and these peaks are the keys to its interpretation. A gamma-ray peak has several notable attributes, as depicted in Figure 8. The most obvious feature is the peak height above background. The centroid, the midpoint of the peak's count distribution, determines the energy of the gamma-ray peak. The energy of a gamma-ray peak identifies the emitting radioisotope, in this instance Cs-137. Hence, determination of gamma-ray peak energies is critical for correct interpretation of spectra. In this report, gamma-ray energies are stated in kiloelectron volts (keV).

The width of a gamma-ray peak, usually measured as the full width at half maximum (FWHM) peak height, is an important measure of gamma-ray detector performance. When an HPGe gamma-ray detector begins to fail, the first sign is often increased peak widths in spectra. In addition, an unusually wide peak may indicate the presence of two adjacent gamma-ray peaks that cannot be resolved by the spectrometer.

Osiris displays the width of the K-40 natural background peak at 1461 keV, permitting operators to check this indicator of spectrometer performance.

By summing the area under the peak and above the background, one determines the net peak area in counts. In turn, the net peak area is proportional to the activity of the emitting radioisotope, as measured in becquerels or microcuries.

3.2.1 Spectral Analysis Logic

The Osiris spectral analysis software supervisory logic is shown in Figure 9. Following initialization and energy calibration, a gamma-ray spectrum is acquired. Next a peak search is performed, and based on the detected and required peaks, the peak-fitting regions are determined. After region-by-region peak fits, the radioisotopes evident in the spectrum are identified by peak energy and the corresponding peak areas and uncertainties noted. For peak detection, Osiris currently requires the peak area to exceed the peak background uncertainty by a factor of 3 or

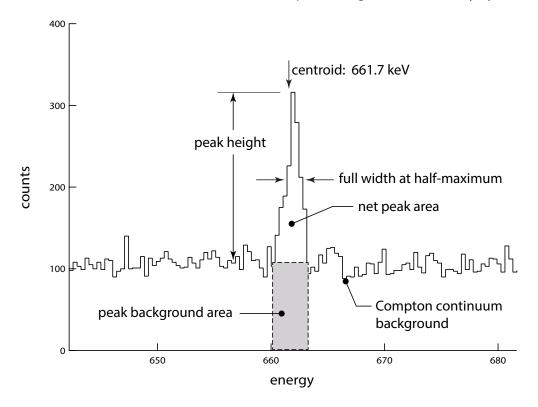


Figure 8. Gamma-ray spectral peak features.

more. Finally, information on the treaty-relevant fission products is pushed across the data filter for display to the user.

3.2.2 Nonlinear Least-Squares Peak Analysis

OSIRIS spectral gamma-ray peaks are characterized with the GAUSS gamma-ray spectrum analysis algorithms of Helmer and McCullagh. [11] GAUSS peak analyses use nonlinear least-squares fits

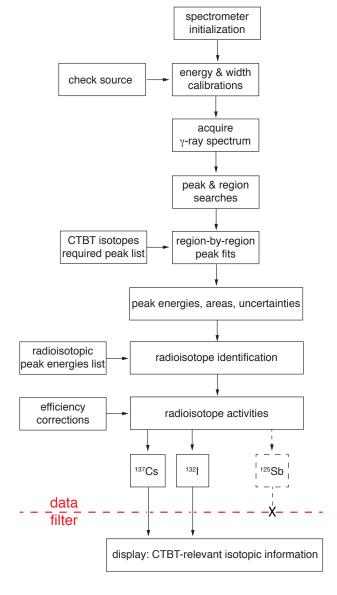


Figure 9. Spectral analysis software supervisory logic

to spectral peaks. This peak analysis method is especially suitable for complex fission-product spectra.

Figure 10 displays a GAUSS fit of overlapping peaks in the 650- to 680-keV region of a nuclear reactor fuel-sample spectrum. Note that GAUSS determines both net peak areas, as indicated by the blue shading of the Cs-137 peak, and the centroid energies, as the arrow in the I-132 peak demonstrates. Also note that GAUSS correctly fits the overlapping I-132 and Sb-125 peaks.

Most of the critical fission-product gamma-ray energies and intensities used in the OSIRIS code are taken from the 1988 textbook of Debertin and Helmer. [12] The isotope half-lives, and the remaining gamma-ray energies and intensities are from the 1996 *Table of Isotopes, Eighth Edition*. [13]

3.2.3 Automated Spectrometer Calibration

The spectrum analysis algorithms rely on accurate calibration of the spectrometer energy scale. For example, the isotope Cs-137 is identified by a gamma-ray spectral peak at 661.7 keV. If the spectrometer energy calibration was just 1% too high, this peak would be misidentified as the 667.7-keV I-132 peak. (Both peaks can be seen in Figure 10.) However, with care, it is possible to calibrate HPGe spectrometers to 0.05% accuracy.

In our experience, energy calibration of a gamma-ray spectrometer can be somewhat difficult for technician-level operators. Hence, we adapted the PINS automatic energy-calibration for use with the OSIRIS system. Using a Eu-152 or Th-228 microcurie-strength check source, a good energy calibration can be achieved in 100 seconds. OSIRIS systems can also be calibrated using natural background gamma rays, but the process requires a 1,000-second count.

A spectrum-blind system like OSIRIS is nearly impossible to energy calibrate, even for experienced spectroscopists, without some type of computer assistance.

The Osiris automatic energy calibration algorithm is described in detail in an earlier report. [14]

3.3 Initial Tests of Osiris Software

The Osiris software was tested with both measured and synthetic fission-product spectra.

3.3.1 Reactor Fuel-Sample Spectra

Seven fission-product spectra from irradiated uranium were analyzed. These spectra were measured from experimental uranium nuclear-reactor fuel samples irradiated in the Idaho National Laboratory (INL) Advanced Test Reactor. The cooling times after irradiation prior to measurement varied from 6 to 24 months. The fission-product gamma-ray peaks are quite intense in these spectra, and since the spectrometer was shielded and tightly collimated, most of the natural background peaks are not evident.

3.3.2 Synthetic Test Spectra

Eight on-site inspection scenarios were defined to design a set of synthetic gamma-ray spectra for OSIRIS data analysis assessment. The scenarios were as follows:

- Underground nuclear explosion strong activity, including particulates release at surface, assayed 4 weeks after explosion.
- 2. Underground nuclear explosion weak activity, with limited particulate release at surface, assayed 4 weeks after explosion.
- Underground nuclear explosion weak activity, with limited particulate release at surface, assayed 1 year after explosion.
- 4. Underground nuclear explosion gas-only release at surface with subsequent decaydaughter particulate deposition, assayed 4 weeks after explosion.

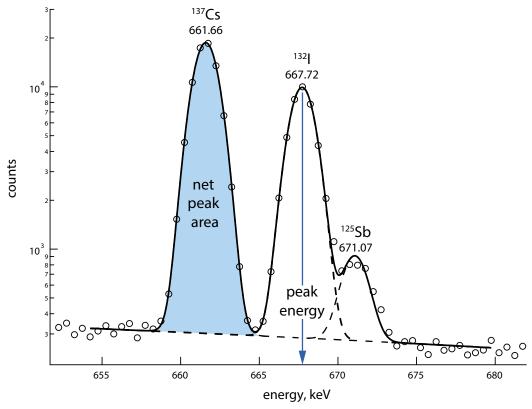


Figure 10. Gauss nonlinear least-squares fit to three fission-product gamma-ray peaks.

- Legacy test-site contamination surface debris and gas seepage assayed decades after explosions.
- Reactor accident predominantly volatile release, like the Fukushima, Japan reactor accident, assayed 3 weeks after the accident.
- Reactor accident core release with refractories & volatiles, akin to the Chernobyl, Ukraine reactor accident, assayed 2 days after the accident.
- 8. Spent-fuel reprocessing waste site radioactivity emissions.

Of these, the first four scenarios are not compliant with the Comprehensive Nuclear-Test-Ban Treaty (CTBT) while the latter four are treaty-compliant.

The synthetic spectra were based on actual measured spectra, to the extent possible, but often computer simulations were necessary to combine the fission-product signal with a natural-background gamma-ray spectrum. As necessary, the synthetic gamma-ray spectra were generated using the Los Alamos MCNP code [15] and the PNNL SYNTH code. [16] In total, this test set contains 100 individual synthetic spectra.

In all cases, the spectra were analyzed twice, once by an experienced spectroscopist using the GAUSS XI interactive gamma-ray spectral analysis program, and once by the OSIRIS automated software. The two analyses were compared and scored by a computer program.

3.3.3 Spectrum Analysis Test Results

The performance of the OSIRIS spectrum analysis software in the tests described above is summarized in Table 3. The OSIRIS spectrum analysis software and the spectrum analysis test are described further in a *Nuclear Instruments and Methods in Physics Research* **A** article. [17]

3.4 Radioactivity Determination

The radioactivity (activity, for short) *A* of an isotope is calculated from the net area *N* of its spectral peak at energy *E*:

$$A = N/(f \cdot \eta(E) \cdot T),$$

where f is the gamma-ray emission fraction, $\eta(E)$ is the absolute detection efficiency at the energy of the gamma-ray peak, and T is the live counting time. The activity calculation is the final step in gamma-ray spectral peak analysis.

The counting time is reported, to millisecond accuracy, by the spectrometer. Each emission fraction is tabulated in reference books like the *Table of Isotopes*.

The detection efficiency of an HPGe detector is a function of the detector crystal size and shape, the counting geometry, and the gamma-ray energy. Since HPGe crystals are not manufactured in standard sizes and shapes like scintillation detectors, their detection efficiency must be calculated or measured as a function of

Table 3: Overall Osiris correct and incorrect isotope identification scores.

Isotope type	True positive fraction, %	True negative fraction, %	False positive fraction, %	False negative fraction, %
Treaty-relevant fission products	98.1	96.2	3.8	1.9
Other fission products	86.5	94.9	5.1	13.5
Other radioisotopes	95.0	96.8	3.2	5.0
Natural background isotopes	83.5	89.7	10.3	16.5

gamma-ray energy for each HPGe spectrometer and each counting geometry used with that spectrometer. Typically, the efficiency of a spectrometer is characterized when it first arrives from the factory. The initial detection efficiency calibration is usually good indefinitely unless the detector is repaired.

For OSIRIS, three geometries are of interest: point-source measurements, *in-situ* geometry counting, and soil-sample measurements.

3.4.1 Point Source Detection Efficiency

The detection efficiency for a point source in a given geometry, e.g., 30-cm from the detector face is determined as follows:

- Measure a calibrated gamma-ray point source in the desired geometry.
- A "mixed" isotope source containing, say, Am-241, Cd-109, Co-57, Co-60, and Y-88 is preferable to a single-isotope source emitting multiple gamma-rays, like Eu-152.

- Determine the net peak areas in the recorded spectrum using a general purpose spectral analysis program like ORTEC MAESTRO. [18]
- Correct the calibration-day activities for the decays of the source isotopes since the source calibration date.
- Calculate the detection efficiency $\eta(E)$ at each mixed-source gamma-ray energy:

 $\eta(E) = N(A \cdot f(E) \cdot T)$, where N is the net peak area, f(E) is the emission probability for a gamma-ray of energy E (some sources emit more than one gamma ray), and T is the measurement live time.

- Fit a polynomial curve through the measured data points. INL uses a cubic-spline curve, calculated with the MATLAB program. [19]
- Provide the efficiency curve coefficients to OSIRIS in a text file.

The detection efficiency curve for a point source 30 cm from an OSIRIS spectrometer front face is shown in Figure 11.

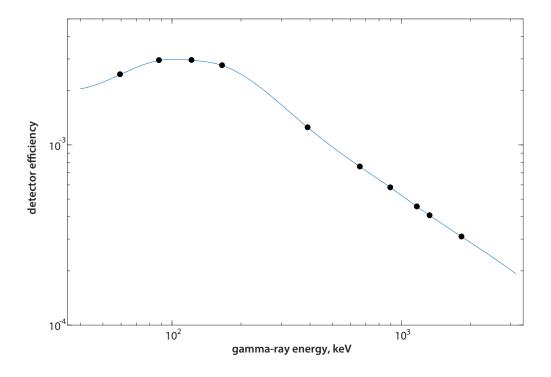


Figure 11. Point-source detection efficiency at 30 cm.

3.4.2 Detection Efficiency for *In-Situ* Counting

The *in-situ* gamma-ray counting method was designed to permit environmental radioactivity measurements over a broad area with little or no need to collect soil samples. [20] In this counting geometry, the HPGe detector is mounted on a tripod, nose down as shown in Figure 12, with the front surface of the HPGe crystal 100 cm above the ground.

A series of detailed Monte Carlo studies, supplemented by gamma-ray measurements, has produced the calibration curve displayed in Figure 13 for an IDM spectrometer used in the *in-situ* geometry. This curve has a 10% uncertainty. Remarkably, the Monte Carlo studies indicate that gamma rays from as far away as 70 meters make a significant contribution to an *in-situ* geometry measurement, and even gamma rays from activity about 500 meters away contribute to the spectrum.

For example, if Cs-137 is distributed uniformly on an infinite flat plane in air (no surface roughness), its 662-keV gamma rays, having have a meanfree-path length of 104 meters (1/e) in air at STP, produce these spectral effects in an IDM-200 spectrometer 100 cm above the ground:



Figure 12. INL and PNNL physicists adjust the height of an OSIRIS spectrometer for in-situ measurements.

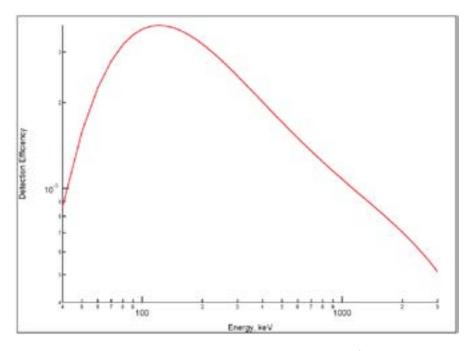


Figure 13. Osiris in-situ geometry detection efficiency.

- 57% of the full-peak count rate comes from inside the 10-meter radius (and 43% from outside 10 meters)
- 90% within a 68-meter radius
- 99% within a 214-meter radius

What maximum radial distance is needed for this Monte Carlo calculation? At 3,000 keV, 99% of the full-peak count rate comes from within a 440-meter radius, probably a sufficient radial distance cut-off in the Monte Carlo calculations. For the *in-situ* efficiency curve as displayed in Figure 13, the solution was calculated out to a radius of 1000 m for 0-3,000 keV. Hence, radial truncation at 1 kilometer produces less than a 1% truncation effect, well within the estimated 10% uncertainty envelope.

3.4.3 Soil-Sample Detection Efficiency

The detection efficiency for gamma-ray peaks depends on a variety of parameters, such as detector size, source location, source composition, and source geometry. Figure 14 shows the modeled peak efficiency for an OSIRIS detector for three different sources, a container with soil in which the radioisotopes distributed uniformly, an empty container with

the radioisotopes distributed uniformly and a point source. The center of all three sources is at the same location, equivalent to 1 cm distance between the nearest sources of the detector and the container. The plot shows the impact of solid angle, going from the point source to the distributed source, as well as the impact of the soil attenuation going from the distributed source to the soil source. These differences motivate a different approach for conducting detector efficiency measurements for a soil sample.

The recommended approach to determine the peak efficiencies for a soil sample is inspired by the semi-empirical approach of Huy et al. [21], which performs an efficiency measurement on a soil sample of known composition and known activity which is then corrected based on radiation transport modeling results. The primary difference between our approach and that of Huy et al., is a simplified correction for energy and density. With this simplified correction, it is estimated that the uncertainty of the calibration can be determined to 4% for all energies at and above 125 keV, and 9% at 88 keV. The primary contribution to this uncertainty is unknown soil composition. Details of the methodology can be found in [22].

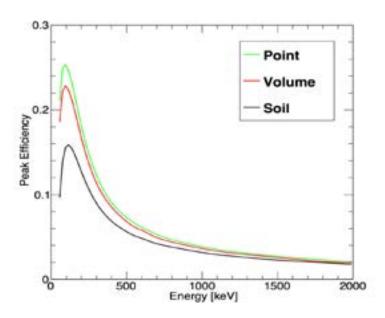


Figure 14. Comparison of peak efficiencies for a variety of sources

The efficiency calibration approach is to determine the efficiency for an unknown soil sample using a measurement on a known soil sample and a correction factor. For a known container at a fixed distance from the detector, the efficiency of an arbitrary soil sample can be determined based on the efficiency of a calibrated soil sample using

$$\varepsilon(\rho, E) = \varepsilon_{c}(\rho_{c}, E)[1 + (0.0827 \ln(E) - 0.6629)(\rho - \rho_{c})])$$

where E is the energy, $\varepsilon(\varepsilon_{c})$ is the efficiency of the (calibration) sample, and $\rho(\rho_{c})$ is the density of the (calibration) sample. This relationship holds only when the calibration soil sample and unknown sample are placed in the same type of container and placed at the same distance from the detector. The initial measurements of the calibrated soil sample can be performed either in advance of the field measurements, or in the field if it is possible to transport the radioactive soil sample to the field counting location.

Our detection efficiency calibration procedure for soil samples is as follows:

- Select a container for the soil samples. The ideal container has a relatively large diameterto-height ratio, with the diameter slightly smaller than the diameter of the detector crystal. The container should be made of low atomic number material to reduce attenuation in the container walls.
- 2. Select a soil sample to use for the calibration sample. Ideally, the calibration soil would be similar to the soil for the unknown sample. The most important variable is the density of the soil. The unknown soil composition introduces a relatively small uncertainty of 4 to 9%.
- 3. Select an aqueous calibrated radioisotopic solution. Such solutions can be found from commercial vendors. One typically needs an activity of a few μCi or less per calibration sample.

- 4. Measure the density of the calibration soil in the container, ρ_{ς} .
- 5. Prepare the calibration soil sample with the radioisotopic solution. Follow appropriate radiation protection procedures. Ensure that the solution is distributed uniformly in the calibration sample and that the container is well-sealed. Allow the solution to evaporate.
- Measure the mass of the calibration sample again to determine if density has changed. It is critical that the density be measured accurately.
- 7. Measure the calibrated soil sample in the desired geometry. It is important that the distance between the detector and the sample be the same for both the calibration and unknown samples; it is especially critical very close to the detector. Set the spectrometer lifetime long enough to acquire 10,000 or more counts in each of the gamma-ray peaks of interest.
- 8. Analyze the gamma-ray spectrum from the source with a general-purpose gamma-ray spectral analysis program like ORTEC MAESTRO [18], and determine the net peak areas of the gamma-ray peaks from the calibration sample, N_i^c. OSIRIS is not suitable for analysis of mixed-isotope calibration source spectra, as it only reports gamma-ray peak areas from the seventeen CTBT fission products.
- 9. Decay-correct the source isotope calibrated activities to obtain the activities A_i , on the measurement date.
- 10. Measure the density of the unknown soil sample, ρ_{c} .
- 11. Determine the detector efficiency $\eta(E_i)$ at each gamma-ray energy E_i by the expression

$$\eta(E_i) = \eta^c(E_i) C(E_i, \rho_s - \rho_c),$$

where the efficiency for the calibration sample is

$$\eta^{c}(E_{i}) = (N_{i}^{c})/(TA_{i}B_{i}),$$

and the correction for the difference in calibration sample and soil sample densities is

$$\eta^{c}(E_{i'}\rho_{s}-\rho_{c}) = 1 + (0.0827 \ln (E_{i}) - 0.6629) (\rho_{s}-\rho_{c}).$$

T is the measurement time and B_i is the gamma-ray emission probability or "branching ratio." Note that for the efficiency measurement, only the density of the soil sample is used as input for the efficiency.

- 12. Apply corrections for coincidence summing if appropriate to the efficiencies. This can be non-trivial, so it is recommended to select isotopes without coincident signatures.
- 13. Fit a polynomial curve through the efficiency points, to obtain the approximate efficiency as a function of energy, $\eta(E_s)$.
- 14. Provide the efficiency curve polynomial coefficients to OSIRIS in a text file.
- 15.Repeat calculation of sample efficiency for each sample with a different density.

3.4.4 Osiris Detection Efficiency Tables

The OSIRIS software includes a set of detection efficiency tables. The current efficiency table is displayed in the top left corner of the main data-acquisition window. Users must ensure that the correct efficiency table has been selected for the measurement they are about to perform.

3.5 Spectral Information for Users

The OSIRIS software filters spectral information measured during an on-site inspection. Only the radioisotopic information needed to conduct the inspection (the "CTBT-relevant radionuclides") is revealed to the user, not the entire gamma-ray spectrum.

3.5.1 Data Filter

A notional spectroscopic measurement data filter [23] is depicted in Figure 15. The data filter hardware and software hide full spectral information from users, revealing only treaty-relevant information on its display device.

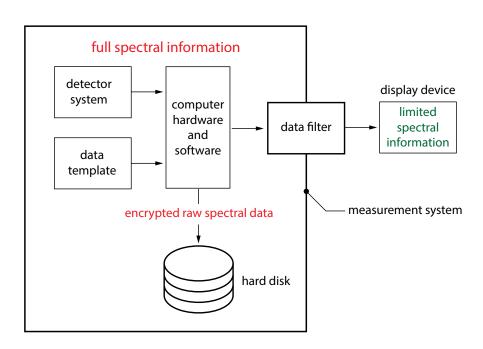


Figure 15. Data filter concept.

3.5.2 Bar-Chart Data Display

The OSIRIS software does not display raw gamma-ray spectra like that shown in Figure 2 to the operator. Instead, the information is limited to radioisotopes relevant to CTBT on-site inspections, and displayed by vertical "intensity bars" as shown in Figure 3. The bar height is proportional to the intensity, in counts/sec, of an isotope's gamma-ray peak.

Osiris also provides information on three natural background isotopes sources to assure users the system is operating correctly.

During a measurement, OSIRIS automatically analyzes the input spectral data, updating its analysis every 10 to 15 seconds. The shading of each intensity bar gets darker as detection confidence for the corresponding isotope increases.

3.5.3 Isotope Information Buttons and Windows

Above the bar-chart display area in Figure 3, there is a series of buttons, one for each CTBT-relevant isotope. Clicking on a button replaces the bar chart with spectral analysis detail for the selected isotope. Spectral information for the fission-product isotope La-140 is shown in Figure 16.

3.5.4 Data Summary Output File

A summary of the spectral analysis is stored for each stored gamma-ray spectrum. Unlike the spectrum, the summary text file is not encrypted. The summary data file for the spectrum displayed in Figure 2, measured from a fresh fission-product source, is shown Tables 4-6.

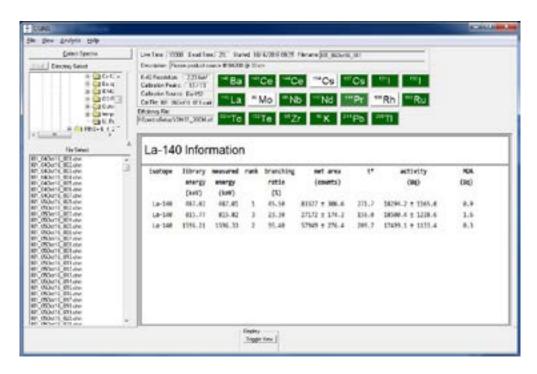


Figure 16. La-140 spectral information window.

Table 4: Summary Spectral-Analysis Text File

OSIRIS Spectrum-Analysis Summary	
File Information	
Spectral Filename	I01_06Oct16_040A
Assay Date and Time	06 October 2016 17:40
Assay Location	PINS Lab
Operator	Jayson Wharton
Public Key File	E:\OSIRIS\fission-product spectra\Cs-134 & Cs-137 spectra\l01_02Jul18public
Sample Description (Header Comments)	Fission product source #104208 @ 30cm
Spectrometer Description	
Spectrometer	IDM-01
HPGe Crystal Temperature	109 (Kelvin)
Live Time (sec)	39000.08
Dead Time	2.40%
K-40 Energy Resolution	2.24 keV FWHM
Analysis Information	
Detector Efficiency File	C:\Program Files (x86)\Osiris 1.7.4\SpectraSetup\IDM01_30CM.eff
Gamma-Ray List	LOCKED_IN_CODE
Program Version	Osiris v1.7.5
Calibration Information	
Calibration File	E:\OSIRIS\fission-product spectra\6 October 16 spectra\101_06Oct16_040.calib
Energy-Cal Date and Time	6 October 16 06:32
Energy-Cal Equation	E(x) = -0.16 + 0.3654x + 0.000e + 00xx (keV)
Energy-Cal Chi Squared	0.019000
Energy-Cal Library Peaks	13
Energy-Cal Peaks Found	13
Engrav Cal Course	Eu-152
Energy-Cal Source	
Max Spectrum Energy	2992.9 (keV)

	Peak	Library	Measured	Branching	Net Peak Area		Activity	MDA
Isotope	Detected?	Energy (keV)	Energy (keV)	Ratio (%)	(counts)	*	(Bg)	(Bg)
Ba-140	Yes	162.67	162.48	6.2	23560.1 ± 221.9	106.2	1.52E+04 ± 1.05E+03	5.30E+00
Ba-140	Yes	537.71	537.26	24.4	31687.7 ± 200.6	157.9	1.58E+04 ± 1.05E+03	1.96E+00
Ce-141	Yes	145.44	145.19	48.2	194971.4 ± 487.2	400.2	1.56E+04 ± 9.76E+02	7.10E-01
Ce-144	Yes	133.54	133.19	11.1	8797.2 ± 200.8	43.8	3.01E+03 ± 2.50E+02	3.24E+00
Cs-134	No	569.32	569.28	15	227.9 ± 88.5	2.6	1.95E+02 ± 8.75E+01	3.18E+00
Cs-134	No	604.69	604.7	97.6	22.2 ± 82.2	0.3	ı	4.70E-01
Cs-134	No	795.84	794.61	85.4	48.0 ± 47.7	10.1	9.73E+01 ± 1.55E+01	4.40E-01
Cs-134	No	801.93	801.93	8.7	49.0 ± 42.8	Ξ:	1	4.29E+00
Cs-137	Yes	661.66	661.47	85.1	682.8 ± 60	11.4	1.18E+02 ± 1.74E+01	4.80E-01
1-131	Yes	364.48	364.54	81.5	10114.9 ± 134.2	75.4	1.02E+03 ± 7.50E+01	4.70E-01
1-131	Yes	636.97	636.7	7.2	481.7 ± 77.7	6.2	9.55E+02 ± 2.11E+02	5.98E+00
1-132	Yes	667.73	667.74	98.7	2331.8 ± 72.3	32.3	3.50E+02 ± 3.18E+01	4.20E-01
I-132	Yes	772.68	772.7	75.6	1608.9 ± 136.1	11.8	3.58E+02 ± 5.17E+01	4.90E-01
I-132	Yes	954.62	954.66	17.8	290.1 ± 43.2	6.7	3.30E+02 ± 6.91E+01	2.38E+00
La-140	Yes	487.02	487.01	45.5	75057.6 ± 291	257.9	1.83E+04 ± 1.17E+03	9.60E-01
La-140	Yes	815.77	815.81	23.3	24524.2 ± 164	149.5	1.86E+04 ± 1.24E+03	1.62E+00
La-140	Yes	1596.21	1596.33	95.4	52101.5 ± 262.3	198.7	1.75E+04 ± 1.14E+03	3.00E-01
Mo-99	No	181.09	181.06	6.1	27.1 ± 158.8	0.2	ı	5.51E+00
Mo-99	No	739.47	739.5	12.1	56.8 ± 45.4	1.3	ı	3.32E+00
Mo-99	No	777.88	777.92	4.3	34.2 ± 60.6	9.0	I	8.65E+00
Nb-95	Yes	765.81	765.82	8.66	36262.9 ± 199.2	182.1	6.06E+03 ± 3.97E+02	3.70E-01
Nd-147	Yes	91.05	90.73	27.9	36036.7 ± 297.4	121.2	4.85E+03 ± 3.31E+02	1.29E+00
Nd-147	Yes	319.41	319.68	2	1480.8 ± 110.8	13.4	5.41E+03 ± 7.29E+02	2.02E+01
Nd-147	Yes	531.02	531	13.1	5342.7 ± 116.4	45.9	4.91E+03 ± 4.02E+02	3.67E+00
Pr-144	Yes	696.51	696.1	1.3	330.5 ± 53.2	6.2	$3.91E+03 \pm 8.63E+02$	3.20E+01
Pr-144	Yes	1489.16	1488.06	0.3	23.5 ± 30.4	0.8	ı	1.30E+02
Rh-106	No	621.8	621.93	10	152.0 ± 68	2.2	ı	4.29E+00
Rh-106	No	1050.4	1050.39	1.6	-40.8 ± 38.4	0	ı	2.90E+01
Ru-103	Yes	497.08	497.07	91	58632.1 ± 260.6	225	7.29E+03 ± 4.70E+02	4.80E-01
Ru-103	Yes	610.33	610.33	5.8	1497.1 ± 123.6	12.1	$3.53E+03 \pm 5.03E+02$	7.98E+00
Tc-99m	Yes	140.51	140.23	100	4358.5 ± 191.4	22.8	1.67E+02 ± 1.74E+01	3.60E-01
Te-132	Yes	228.16	228.3	88	5181.9 ± 150.5	34.4	$2.96E+02 \pm 2.64E+01$	3.90E-01
Zr-95	Yes	724.2	724.21	44.2	32765.3 ± 227.1	144.3	1.18E+04 ± 7.89E+02	9.40E-01
71-95	Ne.	67 237	32 732	7 7 4	7 6 10 10 10 10 10 10 10 10 10 10 10 10 10	000	4 4 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7	

Table 6: Natural Background Isotopic Information					
Isotope	Detected?	Library Energy (keV)	Measured Energy (keV)	Branching Ratio (%)	Net Peak Area (counts)
K-40	Yes	949.83	950.93	15.0	2189.1 ± 107.3
K-40	Yes	1460.83	1460.95	10.7	66713.0 ± 276.3
Pb-214	Yes	242.00	241.92	18.5	10860.6 ± 314.0
Pb-214	Yes	295.22	295.18	18.2	13617.5 ± 265.3
Pb-214	Yes	351.93	351.98	35.8	22093.7 ± 242.5
Tl-208	Yes	583.19	583.11	85.2	14915.7 ± 187.2
Tl-208	Yes	2614.53	2614.56	99.8	13648.8 ± 127.6

3.6 Spectral Data Encryption

To retain and protect sensitive raw gamma-ray spectra, spectral data is encrypted prior to hard-disk storage. Summary analysis information on the seventeen CTBT-relevant isotopes is stored in unencrypted text files. Users have access to the summary spectral information at all times.

OSIRIS encrypts and decrypts spectral data files with the RINJNDAEL block-cipher encryption algorithm of RIJNMEN and DAEMEN. [24] In 2001, this algorithm was selected by the U.S. National Institute of Standards as the Advanced Encryption Standard (AES). [25] OSIRIS employs the AES-128 version using a randomly-generated 128-bit key for encryption and decryption. The AES-128

encryption key, effectively a password, must be safeguarded, and transmitted in an encrypted mode. OSIRIS uses the Rivest-Shamir-Adleman (RSA) public/private key algorithm to generate and securely transmit encryption keys. [26]

When an OSIRIS spectral file has been decrypted, one would like assurance that the decrypted file matches the original spectral file before encryption. OSIRIS computes a hash, a number somewhat akin to a checksum, before encryption and after decryption, and checks that the hash values match. The operator is warned if the hashes do not match. The OSIRIS hash algorithm is Secure Hash Algorithm-256 (SHA-256), designed by the U.S. National Security Agency. [27]

4. Osiris System Integration and Testing

Since gamma-ray spectrometers identify radioisotopes by their associated spectral peak energies, accurate and stable energy calibration is essential for correct isotopic identifications. During an on-site inspection, it is likely Osiris will be used outdoors, not in an air-conditioned lab, and it may not be possible to energy-calibrate the instrument every day. Hence, we have tested Osiris energy-calibration stability versus both temperature and time.

4.1 Energy Calibration Stability vs. Temperature

The OSIRIS spectrometer energy-calibration stability versus temperature was been tested repeatedly in an environmental chamber from $-12\,^{\circ}\text{C}$ ($+10\,^{\circ}\text{F}$) to $50\,^{\circ}\text{C}$ ($122\,^{\circ}\text{F}$). The variations of the Eu-152 121-keV and 1408-keV peak-centroid positions vs. temperature, typically less than \pm 0.21 keV, are shown in Figure 17.

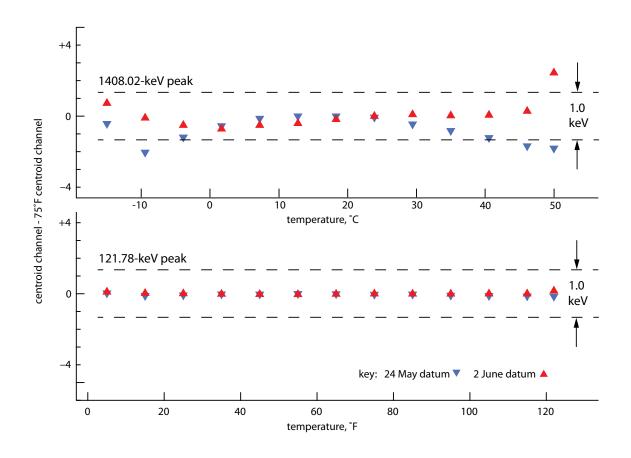


Figure 17. Energy calibration stability vs. temperature.

4.2 Energy-Calibration Stability vs. Time

If a spectrometer is energy-calibrated daily, its calibration must remain stable for at least 24 hours. As a baseline, we first monitored the spectrometer energy-calibration stability indoors, and found it remained stable over 5 days within 0.031 keV (1 σ). Next, we conducted month-long tests of the spectrometer outdoors at both Idaho National Laboratory (INL) and Pacific Northwest National Laboratory (PNNL). Testing at INL took place during winter conditions, as shown in Figure 18. The INL winter energy-calibration stability test results are displayed in Figure 19, and the PNNL summer test results are shown in Figure 20. The system energy calibration remained constant within \pm 0.113 keV in both tests.

4.3 Cs-134/Cs-137 Ratio

The ratio of Cs-134 to Cs-137 activity helps differentiate between fission-product spectra produced by a nuclear weapons test, and spectra produced by a nuclear reactor accident or by medical isotope processing. Nuclear explosions do not produce Cs-134, and hence the Cs-134/Cs-137 activity ratio is zero in fission-product spectra measured after a bomb test. In a nuclear reactor, Cs-134 is produced by thermal neutron capture on stable Cs-133. For example, following the Fukushima Daiichi accident, the Cs-134/Cs-137 ratio was approximately 1.0. [28]



Figure 18. Winter testing of OSIRIS at INL

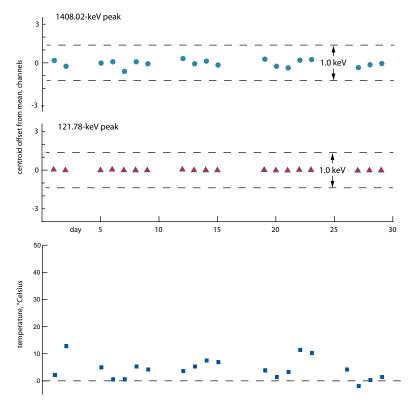


Figure 19. Winter energy calibration stability vs. time.

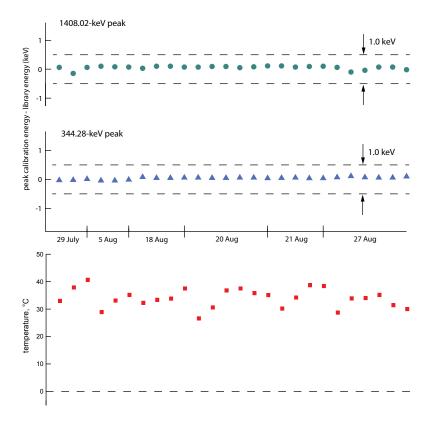


Figure 20. Summer energy calibration stability vs. time.

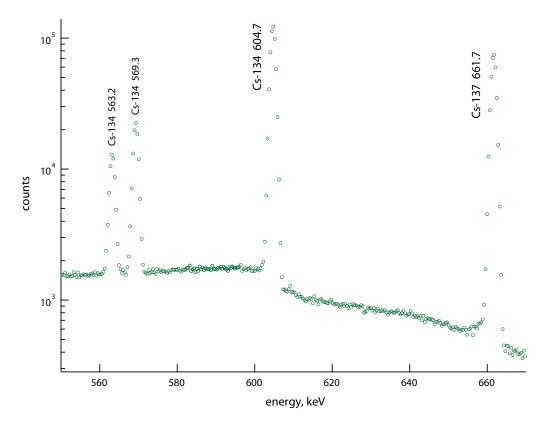


Figure 21. Cs-134 and Cs-137 spectral peaks.

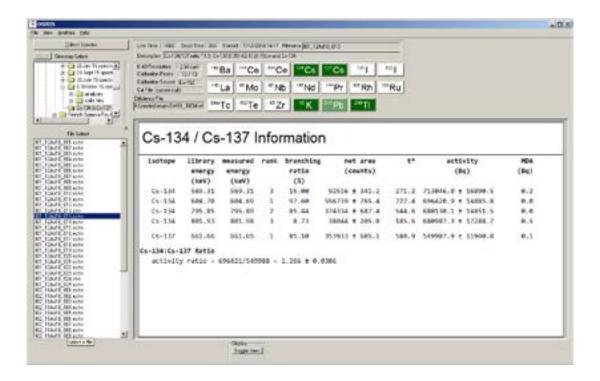


Figure 22. Cs-134/Cs-137 spectral information window.

Since the Eckert & Ziegler commercial fission product sources do not contain Cs-134, we have used individual Cs-134 and Cs-137 sources to verify that the activities of those isotopes is quantified correctly. An example Cs-134 and Cs-137 spectrum is shown in Figure 21, and the corresponding Cs-134/Cs-137 spectral information box, which includes the Cs-134/Cs-137 activity ratio, is displayed in Figure 22.

4.4 Fission-Product Identification Amid Nevada Test Site Legacy Radioactivity

In March 2017, a field test of OSIRIS was conducted in two areas at the Nevada National Security Site, formerly the Nevada Test Site. Two OSIRIS instruments were operated in the *in-situ* (downward-looking) counting mode, and the radioactive background in one test area was a

factor of 15 higher than in the other test area. A six-months-old fission-product source was placed 1 meter directly below the spectrometer during measurements, as shown in Figure 23. It was removed during background runs. While many of the short-lived isotopes in the source had decayed away, Osiris detected two CTBT-relevant fission products: 64-day Zr-95 and its 35-day daughter, Nb-95 at both test areas.

4.4.1 Unscheduled Drop Test

On the last day of testing in Nevada, a storm developed suddenly, and the related gale-force winds tossed the plastic shipping container where the notebook computer was being sheltered. Alas, the computer-spectrometer USB cable yanked over the spectrometer on its tripod, causing it to fall 1 meter (40 inches) to the ground. The spectrometer was sent to the ORTEC



Figure 23. Osiris in-situ measurement in progress at Yucca Flat on the Nevada National Security Site, formerly the Nevada Test Site.

factory for evaluation, and it was found to meet its original specifications. In short, it survived a 1-meter drop without damage, in keeping with the applicable standard for portable radiation detection instrument ruggedness. [29].

4.5 Fission-Product identification vs. Time

OSIRIS is designed to measure fission-product isotopes during an on-site inspection under the CTBT. To evaluate OSIRIS measurement accuracy, we recorded gamma-ray spectra from a fission-product radioactive source as it decayed. Our evaluation criterion is simple: do the OSIRIS spectroscopic data points match the known decay curves of fission-product isotopes?

We used a commercial 3-microcurie (0.12-megabecquerel) mixed-fission-product source containing all of the CTBT-relevant isotopes, save Cs-134 and Rh-106. [30] When new, this source produced the gamma-ray spectrum in Figure 2. Osiris counted the source almost every day, in order to follow fission-product decay for nearly one year at INL. Decay curves were calculated from fission-product half-lives, not from a least-squares fit to the experimental data. For Nb-95, the daughter isotope of Zr-95, the calculated curve includes parent-daughter effects. The decay curves for six representative CTBT-relevant isotopes (solid black lines) are compared to the measured spectral data (small colored circles) in Figure 24, and the agreement between the theoretical decay curves and the experimental data points is excellent.

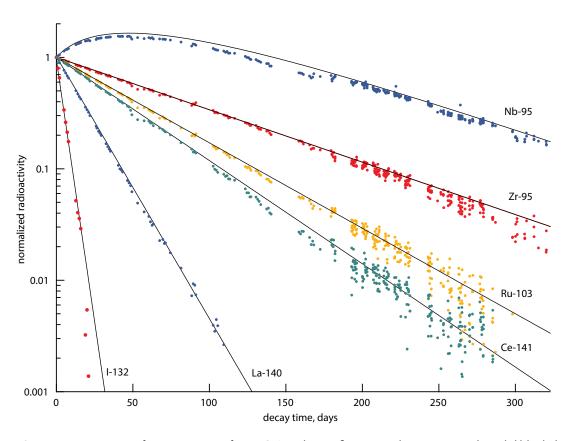


Figure 24. OSIRIS measurements of activity vs time for six CTBT-relevant fission-product isotopes. The solid black decay curves are calculated from fission-product half-lives, and the colored circles are experimental data points.

5. Conclusions

The OSIRIS hardware and software has been specifically designed for the acquisition and analysis of high-resolution gamma-ray spectra during on-site inspections under the Comprehensive Nuclear-Test-Ban Treaty (CTBT).

Starting in 2014, extensive tests of Osiris have been conducted, to validate its design and verify its performance with fission-product measurements and data analysis.

5.1 Design Summary

The OSIRIS system hardware includes an ORTEC IDM-200-V high-purity germanium spectrometer, controlled by a ruggedized notebook computer running custom data acquisition and data analysis software. Similar instruments have been used by the U.S. military for over ten years.

The OSIRIS software design rests on code used for over twenty years in U.S. military field operations. The key software feature of the OSIRIS system is a data filter that limits its display to only radionuclide information relevant to CTBT on-site inspections.

5.2 Testing Summary

The OSIRIS gamma-ray spectrometer, then an ORTEC Transpec-100, was tested successfully at the 2014 Integrated Field Exercise (IFE14) in Jordan. Subsequently, the Transpec-100 was replaced by an ORTEC IDM-200-V spectrometer, for data security reasons.

A spectrum-blind instrument like OSIRIS must remain stable between energy calibrations, and hence we have tested the OSIRIS energy-calibration stability versus time and temperature. In temperature tests over a 62 °C (112 °F) range, the OSIRIS calibration remained stable within 0.23 keV. It is unlikely a spectrometer would encounter

daily temperature changes this large during an on-site inspection. OSIRIS energy-calibration stability versus time was evaluated in month-long outdoor tests, in winter at INL and in summer at PNNL, and in these tests, the calibration remained stable within 0.11 keV.

Recently, OSIRIS was field-tested at the Nevada National Security Site. It identified two shortlived CTBT-relevant fission products amid legacy radioactive backgrounds, plus it survived an unplanned 1-meter drop test.

To characterize OSIRIS identification of CTBT-relevant radioisotopes, we measured gamma-ray spectra from a calibrated uranium fission-product source for almost one year. As the decay curves in Figure 13 attest, OSIRIS has correctly measured CTBT-relevant fission-product isotopes as their activities decayed by three orders of magnitude.

5.3 Commercialization and Technical Readiness Level

Osiris has recently been licensed to the ORTEC Division of AMETEK, Inc. To this end, we have designed custom shipping boxes for the hardware; tested a set of user-training-class materials, written a 100-page User's Manual [31], and created a software distribution/installation disk.

Given its extensive system testing and recent commercialization, the instrument now qualifies as Technical Readiness Level 8 (TRL-8).

Acknowledgments

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